

Sustainable Utilization of Agricultural Straw for Harmful Algal Blooms Control: A Review

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Abstract: The use of agricultural straw for algal bloom control has been studied for more than 30 years. In this article, we have reviewed the promising potentials of using agricultural straw as source of anti-algal agents, including the effectiveness of each major straw type so far used in this regard, and the investigated algal species. Various pre-treatment methods have also been widely reviewed. Significant progress has been made in natural product chemistry and molecular biology with regards to agricultural straw, especially in relation to the extraction of anti-algal allelochemicals, degradation processes of agricultural straws and the mechanisms through which these inhibitions occur. The development of biotechnologies using agricultural straw to successfully inhibit growth of bloom forming algae has been generally accepted as environmentally friendly. The current research status and that of the future should include isolation and discovery of antialgal allelochemicals, development of models that would illustrate the sequence of physiologic events that match the species-specific inhibitor phenomenon, and products fit in the field applications.

Keywords: Agricultural straw; eutrophication; harmful algal blooms; allelochemicals; algal inhibition; bioremediation

1 Introduction

Algal blooms would occur in any water body when the environmental conditions are favorable (i.e., eutrophic water body) [1–3]. Harmful algal blooms (HABs) may result from explosive growth of either an algal species that dominates the water column rapidly, or highly toxic species even if it does not accumulate in extremely high numbers [4–7]. In some cases, toxic conditions occur even when the water is clear and with low algal cell concentrations [8]. The bloom of cyanobacterium *Microcystis aeruginosa*, for instance, is a ubiquitous phenomenon in eutrophic lakes and reservoirs in many countries [9,10]. Many strains of *Microcystis* are known to produce cyanobacterial hepatotoxins called microcystins [11]. The toxin, a soluble peptide, is poisonous to aquatic organisms and damages zooplankton, fish, and the



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liver of higher organisms including human beings [12]. Throughout history, blooms of microscopic algae have had a major impact on fish, birds, mammals and other organisms in the marine food web [13]. The algae grow quickly and form thick layer, becoming difficult to treat.

Large amounts of agricultural waste are produced annually at the same time. According to United Nations food and agriculture statistics database, the production of cereals and crops in the past ten years (2008–2017) constitute about 31 billion tons (Fig. 1) [14]. The nine most prominent crop types constitute 96.4% of the total grain, including peanut (*Arachis* spp.), canola (*Brassica* spp.), cotton (*Gossypium* spp.), rice (*Oryza* spp.), wheat (*Triticum* spp.), maize (*Zea* spp.), soybean (*Glycine* spp.) [15]. Others include barley (*Hordeum* spp.), sorghum (*Sorghum* spp.), millet (*Panicum* spp.) and rye (*Leymus* spp.). For instance, China alone being one of the largest agricultural countries in the world produced a total crop yield and sown area amounting to 657.89 million tons and 117.04 million hectares in 2018, respectively, making it one of the most abundant countries in terms of straw resources in the world [16]. Agricultural residues are generated as a by-product of certain crops and are among the most abundant agricultural by-products in the world [17], also the largest clean renewable and sustainable resource on the planet.

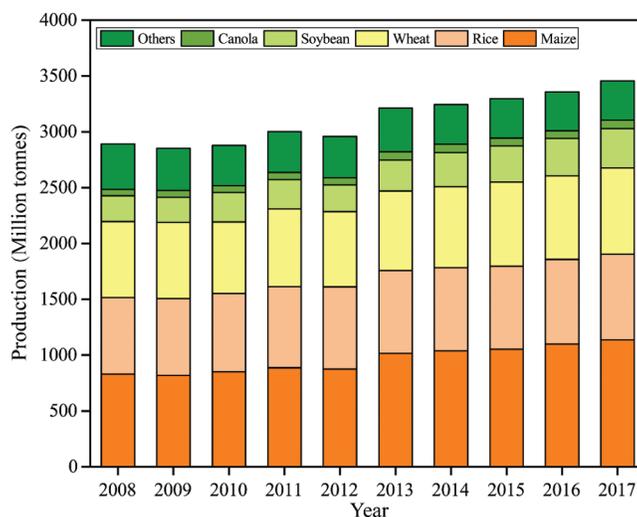


Figure 1: Global cereals/crops production for 2008–2017, including canola, soybean, wheat, rice maize, and others (barley, sorghum, millet, rye, etc.) (data source: FAO) [14]

After decades of research and development, straw utilization techniques are becoming more practical and economical. As an integral part of the bio-circular economy, straw utilization leads to energy saving and reduced emission of air pollutants [18]. Currently the economic, bio-resource and environmental benefits of straw usage is a common knowledge. However, despite the many usages of straw, like sugar, fuel, fertilizer, domestic animal feed, and material for furniture, etc., these wastes are disposed of indiscriminately, especially in developing and under-developed countries. One of the major constraints is the huge difference in the farmers' awareness levels toward straw utilization in different areas [19,20]. In addition, the hidden costs in straw utilization process often prevent its promotion. For instance, in developed countries like the USA and Canada, straws are used as raw material for biofuels [21]. The major drawback to this approach is the cost of straw transportation from farms to the bio-refinery [22]. Therefore, some farmers resort to directly burning the straw on the field, constituting a serious air pollution. Estimation of emissions from open-field straw burning has been carried out at national levels using emission factors and quantity of straw burned, e.g., in India, Thailand, the Philippines, US, and China [18].

Therefore, expanding the use of low or non-existent environmental impact materials such as agricultural straw in the field of algal inhibitions is urgent and meaningful, which fits into a context of effective bioremediation and sustainable development. However, researches should be geared towards technological advancement, implementation of environmental policies and education. Actually, aquatic macrophytes, which are quite similar to agricultural straw, have been widely used to inhibit HABs [23–26].

With this review we aim at providing an overview of the sustainable utilization of agricultural straw in HABs control, with specific objectives to (1) analyze existing studies on algal inhibition by agricultural straw, (2) present methods of straw pretreatment to prepare anti-algal materials, (3) summarize the discovered key allelochemicals; (4) illustrate the algal inhibiting mechanism of agricultural straw and its products, and (5) consider the advantages and disadvantages of practical applications.

2 The Potential and Researches of Using Agricultural Straw as Source of Antialgal Agents

Plants are rich in active compounds or secondary metabolites such as alkaloids, steroids, tannins, glycosides, volatile oils, fixed oils, resins, phenols and flavonoids. These compounds are present in their organs such as leaves, flowers, bark, seeds, fruits, root, etc. Extraction processes of these metabolites are related to the difference in solubility of the compounds present in a mixture of solvent [27,28]. The beneficial action of those plant-constituents typically come from the merging work of these secondary metabolites [29]. Agricultural residues such as barley straw [30–33] has been used extensively and has shown some significant potentials to be an effective agent in controlling algal blooms in both freshwater and marine ecosystems. Its ability to inhibit a variety of species at reasonable straw concentrations has been investigated at rivers, ponds, canals, reservoir and coastal water levels.

2.1 Agricultural Straws That Have Been Tested

In the past years, six types of straws have been applied for inhibiting harmful algae, predominantly barley straw, rice straw and wheat straw respectively. Other agricultural straws proven to be effective on HABs inhibition include corn straw, canola straw and soybean straw.

Our investigation revealed that barley, rice, corn and wheat straws have been used in 131 cases with 100 cases success, 112 with 88 cases success, 12 with 8 cases success, 10 with 7 cases success, representing 76.34%, 78.57%, 66.67% and 70.00% success rate, respectively. Soybeans (pod) and canola straw achieved 100% inhibition rate in all the investigations, respectively, and the individual experimental results showed no failure (Fig. 2). Some of the failed cases have been traced to inappropriate pre-treatment methods. For instance, pre-treated rotted straw succeeded but the autoclaved straw failed [34,35]. Waybright et al. used different solvents for straw extraction, most of the investigated organic solvent extracts were successful in algal inhibition, but the ethanol extract failed [36]. Sometimes, filtration also contribute to the anti-algal effect, i.e., the use of filtered decomposition failed, but the use of unfiltered extract succeeded [37–39]. Different straw types was another reason found, for instance, the use of barley straw was highly successful as against wheat straw that failed in an experiment conducted by Ball et al. [40]. Extract concentration was yet another reason. Certain concentration were found to inhibit while others stimulated algal growth, for instance concentrations between 10 g/L to 30 g/L for rice straw inhibited the algae while lower concentrations (< 10 g/L) stimulated algal growth [41,42].

2.2 Key Cases of Straw Application

2.2.1 Barley Straw

It's the first species of agricultural residues proven to be effective on HABs inhibition, and there has been steady investigations showing a growing body of evidence that decomposing barley straw could inhibit algal growth. It has also been the only species used successfully in the field applications. For instance, the Tibetan barley straw was found as an algistatic agent, effectively inhibiting algae at concentrations of 2.0 to 8.0 g/L

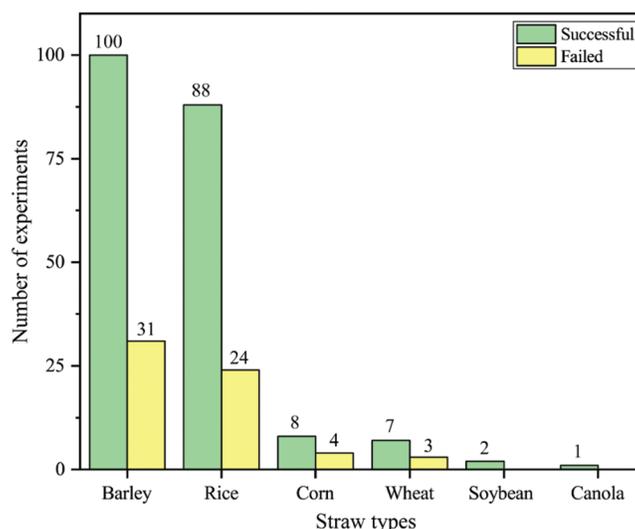


Figure 2: Number of studies on algal inhibition using agricultural straws for the six major types of agricultural straws

[43]. Finely chopped barley straw has been found to produce 90% inhibition of algal growth at the dosage of 4 g/L in a reservoir [32,44]. Barely straw deployment limited summer growth of algal populations in Lake Williston, a recreational lake in Maryland, USA [45]. Growth of *Cladophora glomerata* in Chesterfield canal has been found to show a significant inhibition when barley straw extract was applied [31]. The presence of rotting barley-straw at approximately 50 g m⁻³ in a disused water supply reservoir has been seen to significantly reduce both the general phytoplankton productivity and the cyanobacterial dominance [32]. In 1993 and 1994, treatment with barley straw on a potable supply reservoir with a long history of diatom and cyanobacterial blooms in spring and summer respectively, did not only notice a reduction in blooms but also reduced the processing costs and improves the efficiency of the treatment process enabling other pathogens to be removed more effectively [46]. At the Royal Canal where filamentous algae continuously interfered with amenity exploitation and water management, bales of barley straw were used to reduce dense growths of filamentous algae [33]. A research on long-term algal control in a reservoir using barley straw showed a continuous suppression on populations of cyanobacteria, diatoms and unicellular green algae [47].

A simple aqueous extract prepared from decomposed-barley straw was found to inhibit the cyanobacteria *Microcystis* sp., at a very dilute concentration (0.005%) [40]. Although there have been limited attempts to control harmful algae in coastal and estuarine waters, inhibition of some dinoflagellate species (*Gyrodinium galatheanum*, *Gymnodinium sanguineum*, *Heterocapsa triquetra* and *H. pygmaea*) has been successful using barley straw [48]. A new record that reveals the susceptibility of freshwater and brackish phytoplankton taxa to barley straw extracts including species-specific responses and shifts in species dominance in mixed assemblages has been provided [49]. Purcell et al. [50], on their comparative study of five site within UK water companies using barley straw observed that straw was selectively inhibitory towards specific algal groups, these and many more investigations on the potency of barley straw extract have been tested and proven to be effective. Barley straw has been found to contain some toxicologically relevant levels of phenols and oxidized phenolics derived from lignin as the degradation of straw occurs [44]. The extraction of salcolin A & B and their inhibitory effect of *M. aeruginosa* were discovered in barley straw [27]. These substances had different physiological effects on algae [27].

2.2.2 Rice Straw

Though rice extract has not been used as widely as barley straw, investigations show that it also has a good successful ratio. Rice straw extracts at 0.01 mg/L was found at 98% inhibition rate on *M. aeruginosa* on day 8 [51]. Investigation was also carried out on algicidal effects of rice hull extracts on the growth of *M. aeruginosa*, at 1000 µg/L with a 64% inhibition rate [52]. A recent study where rice and rye straws were used on the growth of *M. aeruginosa*, clearly showed that both extracts inhibited the cyanobacteria [53]. In a continuous culture system, rice straw aqueous extract at 10.0 g/L was observed to suppress the growth of *M. aeruginosa* with a 98% inhibition rate after 9 days of treatment [54]. Investigation on the physiologic and morphologic responses of cyanobacteria to rice straw extract revealed that the cell membrane potential, cell size, and in vivo chlorophyll-a fluorescence were significantly affected [55].

2.2.3 Wheat Straw

Wheat straw in algal inhibition experiments has been scanty. In an experiment to clarify the inhibitory mechanism of wheat bran leachate (WBL) on *M. aeruginosa* [56], the extract has shown to have a compelling inhibitory effect on *M. aeruginosa* with a concentration equivalent to 1.6 g dry mass of wheat bran per liter. Results also revealed how the photosynthetic systems and membranes were the potential targets of toxicity of WBL on *M. aeruginosa*, and the oxidative damage being an important mechanism explaining the inhibitory effect of WBL on *M. aeruginosa*. An attempt was made to extract valuable compounds from wheat straw degraded by a fungi *Pleurotus ostreatus* [22], most of these extract were from phenolic compounds that have been implicated in the inhibition of algal blooms.

2.2.4 Other Straws

Other agricultural straws used for HABs control include corn straw, canola straw and soybean straw. For instance, Yang et al. [57] confirmed the growth inhibition of corn straw and corn leaf on *Alexandrium tamarense*, the extracts in GC-MS result revealed that the fatty acids might be responsible for the inhibition. Wang et al. [58] studied the inhibitory effect of different agricultural wastes on *M. aeruginosa* including barely straw, wheat straw, rice straw to soybean straw, soybean pod, corn straw and canola straw. All the wastes could work and soybean pod achieved the highest inhibition rate.

2.3 Algal Species Tested with Straw Extract Effect

We investigated algal species used in HABs inhibition by agricultural straw for the past three decades. Out of the totally 201 cases in fresh water, 154 was successful while there were 47 failed cases, representing 76.62% success rate. In coastal water, 67 cases were recorded, 52 were successful and 15 failed cases occurred, showing 77.61% success rate. Interestingly, the ratio of success in the fresh waters are similar to that of coastal waters, although 56.9% of the studies have been performed in the fresh waters while 43.1% have been carried out in coastal waters.

The algal species tested for susceptibility to straw extract have been documented and divided by their phylogeny categories (Fig. 3). Phytoplankton species of the phylum Cyanophyta, Chlorophyta and Chromophyta appeared to be the most successfully investigated. In phylum cyanophyta, out of 115 experiments recorded, 93 succeeded and 22 failed, representing success rate of 80.9%. *M. aeruginosa* and *Nostoc* sp., were the most successful, while mostly failed was *Anabaena* sp. [59]. For phylum chlorophyta, 49 were successful while 20 failed in 69 experiments, representing 71.0% success rate dominated by *Scenedesmus* sp., and *Cladophora glomerata* while some failed species include *Chlorella vulgaris* and *Selenastrum capricornutum* FACHB-271 [60]. In total, 47 cases were captured in the phylum Chromophyta, 42 succeeded and 5 failed, showing 89% success rate mostly of *Phaeocystis globosa*. In the phylum Pyrrophyta, 20 cases were recorded, 10 succeeded and 10 failed, representing 50% success rate, *Alexandrium tamarense*, *Gyrodinium galatheanum* and *Heterocapsa triquetra* all responded, among the failed were *Gyrodinium estuariale*, *Peridinium* sp., and *Ceratium furca*. The

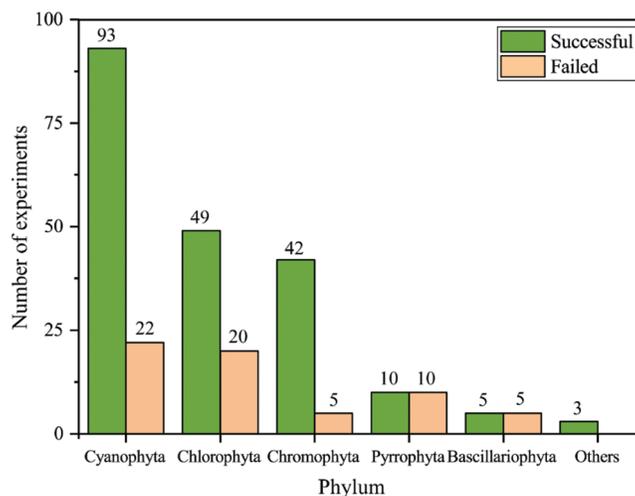


Figure 3: Number of studies on algal inhibition using agricultural straws for the major phytoplankton phylum

phylum Bascillariophyta recorded 5 success and 5 failure cases, successful results were found in *Tabellaria flocculosa*, *Asterionella* sp. and *Melosira* sp. while *Navicula* sp., *Cyclotella* sp. and *Synedra* sp. failed [61]. Inhibition of diatoms, dinoflagellates and others have not been widely documented. Phytoplankton species showed high species-specific responses to agricultural straws. Rice straw, for instance, successfully inhibited *Anabaena* sp. and *Nostoc* sp., but stimulated the growth of *Chlorella* sp. [59]. Corn leave extract stimulated the growth of *P. globosa* but inhibited several dinoflagellates [62]. Barley straw stimulated diatoms and the cyanobacteria *Oscillatoria* sp. while it inhibited various green algae, and other cyanobacteria *Microcystis* and *Anabaena* sp. [63,64].

3 Pre-treatment Approaches

Plant cell wall constitute of cellulose, hemicellulose and lignin. Pre-treatment of straw is needed to break the block of cell wall for degradation and release of allelochemicals into the environment. Pre-treatment is required to increases surface area available for enzymatic hydrolysis during degradation, alter the biomass particle size and structure as well as its sub-microscopic chemical composition. In addition, pretreatment will benefit breaking the structure so that hydrolysis of the carbohydrate fraction to monomeric sugars can be achieved rapidly with greater yields [65]. In applying straw directly in the field, the general pre-treatment methods include drying and packaging them in bales and releasing them to water bodies [66,67]. Other investigators tried to physically crush the straws, sieve and use them directly to inhibit algae [42,68], which have also produced good results. The most frequently used approach in straw pre-treatment is chopping them into small particles, and then soaking in distilled water and obtaining the straw extract. It is considered as the critical step and has a large impact on digestibility of cellulose, and strongly affect downstream costs involving detoxification, enzyme loading, waste treatment demands [69].

Combinations of approaches have also been used to ascertain the most effective method of pre-treatment. Waybright et al. attempted three methods; for sample 1, decomposing dried, chopped barley straw (about 2 cm lengths) was prepared in 5 L deionized water, air was introduced, for 154 days at room temperature. Sample 2, extraction was done using dichloromethane/methanol (1:1) for 18 h. The organic extract was dried and stored for further testing. For sample 3, extraction was done with dichloromethane/methanol (1:1), the extract dried, and the straw soaked overnight in water at room temperature [36]. All three aqueous extracts were found to show similar inhibitory effects against *M. aeruginosa* at concentrations of 1.72, 2.40 and 3.03 g/L respectively [36]. The compounds responsible for the algistatic activity of the straw were observed to release in bits over a period of time, but not at once in a short

time. However, sample 2, soaked for approximately 1 month, showed the same algistatic profile as the samples prepared at 2 days and 157 days. This suggests that the class of compounds responsible for the algistatic properties are polar molecules (water soluble) released as early as 1 day after introducing the straw into an aqueous system, and are not derived from photochemical or microbial processes.

Phenols derived from lignin in straws are difficult to release under natural conditions, however, microorganisms such as white rot fungi is capable to enhance the degradation of straws, reduce the degradation time from 8 weeks for fresh straw to 1 week for rotted straw [70]. During fungal pre-treatment, there are no alternative sources of nitrogen, this leads to greater bacterial decomposition of straw lignin to inhibitory substances. The efficiency of pre-treatment depends on the chemical composition, physical structure of the agricultural straw biomass and the treatment requirement [71]. Although each of the pre-treatment approaches result in its unique minimum concentrations, none of the approaches so far developed have been reported to have any detrimental effect on the system. Pre-treatment of agricultural straws for algal inhibition purposes either by physical, chemical, biological or synthesized approaches have been successful and has contributed to the inhibitive effect of straws (Tab. 1).

4 Anti-algal Allelochemicals in Agricultural Straw

The phenomenon of allelopathy encompasses all types of chemical interactions among plants and microorganisms. Several hundred different organic compounds (allelochemicals) released from plants and microbes are known to affect the growth or aspects of function of the receiving species [90]. It has been seen as a new weapon against invading toxic species, and has become a hot topic in ecology in recent years [27,28,91]. The debate of whether the algal inhibitors were released from decomposing barley straw or were of microbial origin was put to rest when Irene Ridge et al. [73] worked on towards understanding the nature of algal inhibitors from barley straw. The inhibitor is believed to originate from the oxidation of phenolic extracts released from aerobically decomposing straw, for instance, barley straw has been found to contain ester compound (1,2-benzenedicarboxylic acid) tested to be a major anti-algal chemical [78], also rice straw extracts [51] have been tested to contain an allelochemical (salicylic acid), found at different concentrations to inhibit algae. One of the latest findings have shown that a pair of chiral flavonolignans isolated from barley straw extract using a bioassay-guided isolation procedure against *Microcystis* sp. contained salcolin which is the key allelochemical in the straw, effective in inhibition of cyanobacteria [27].

Many studies have focused on the separation, identification, release pathways, application potential, including functional mechanisms of allelopathic materials, and much progress has been made [91–94]. Inhibition by barley straw has been proven to be associated with solubilization and oxidation of lignin. From our investigation, it is revealed that barley straw has the widest range of inhibitive allelochemicals (Tab. 2). These allelochemicals can be grouped into different classes of natural compounds as seen in Tab. 2. So far, there has been no particular group of compounds traced to having specific algicidal or algistatic effects on specific algal species. Allelopathic compounds like Benzoic acid, Vanillic acid, Syringic acid and *p*-Coumaric acid are found in both rice and barley straws.

Allelopathic algicides are as potential natural resources found to deter exclusive and efficient characteristics HABs in both fresh and coastal waters. Several field applications show that agricultural straws are important to maintain a clear and bloom free water regime within a water body [33,46,66,74]. Although excessive application of agricultural straw to a water body may cause a shift of regime from clear water to turbid water. Apart from the release of allelochemicals for algal inhibition, moored barley bales have also been found to provide useful substrate for benthic invertebrates. The benthos act as detritus and nutrient trap, and would thus help reduce the algal growth [67]. In addition, identifying allelochemicals would also help us to make selections on the specific crop type in the geospatial planning of the landscape plant in rural and urban aquatic environment. According to the phylogeny and nature of

Table 1: Pre-treatment methods of agricultural straws and effective minimum algal inhibition concentration

Category	Straws type	Target algae	Straw pre-treatment method	Minimum effective concentration	References
Direct use	barley	<i>Microcystis aeruginosa</i> , <i>Anabaena</i> sp., <i>Aphanizomena</i> sp.	straw bales packed in nets containing 2 kg barley and soaked in water	40.0 g/m ² DW	[64]
	barley	<i>Anabaena</i> sp., <i>Asterionella</i> sp.	7 tones introduced into reservoir inlet	worked but no specific concentration	[46]
	barley	<i>Cladophora glomerata</i>	20 bales of 20 kg straw soaked in canal	worked but no specific concentration	[66]
	barley	<i>Cosmarium</i> sp.	7 bales placed around shoreline	worked but no specific concentration	[67]
	barley	<i>Oscillatoria tenuis</i>	3.5 tones straw in six floated booms at reservoir surface for 4 months	25.0 g/m ³ DW	[44]
	barley	<i>Anabaena</i> sp., <i>Asterionella</i> sp., <i>Stephanodiscus</i> sp., <i>Synedra</i> sp., <i>Tebellaria</i> sp., <i>Melosira</i> sp.	1.5 tones straw was put in netting system and attached to floats	6.0 g/m ³ DW	[47]
	barley	<i>Cladophora glomerata</i>	bales of straw anchored along the canal banks at 50 m intervals	113.0 g/m ² DW	[33]
	barley	<i>Microcystis</i> sp.	straw dosed in four floating lines near river inlet to cover 33% area	5.0 g/m ² DW	[50]
	barley	<i>Anabaena</i> sp., <i>Apanizomenon</i> sp., <i>Apanothece</i> sp. & <i>Planktothrix agardhii</i>	150 bales of straw attached to five anchorage floating structures.	50.0 g/m ² DW	[72]
	barley	<i>Microcystis aeruginosa</i>	500 bales of straw were deployed at feeder streams and lake.	worked but no specific concentration	[45]
Crushed	barley	<i>Phaeocystis globosa</i>	chopped into different size: $\varphi = 2 \text{ cm}/\varphi$ (0.45–0.90) mm/ φ (0.45–0.30) mm/ φ (0.09–0.30) mm/ $\varphi \leq 0.09$ mm	worked but concentration not record	[42]
	barley	<i>Cryptomonas</i> sp.	straw chopped coarsely (5 cm)	4.0 g/L DW	[32]
	barley	<i>Microcystis aeruginosa</i>	straws chopped finely to (1.4 mm) at 18–25°C for 4 weeks	10.0 g straw/L unrotted	[73]
	barley	<i>Microcystis aeruginosa</i>	by decomposing 50.3 g dried, chopped pieces (2 cm) straw for 154 days at room temp	10.1 g/L DW	[36]
	barley, canola, corn, wheat, soybean, rice	<i>Microcystis aeruginosa</i> FACHB-915	crushed into particles	2.0 g/L DW	[58]

Table 1 (continued).

Category	Straws type	Target algae	Straw pre-treatment method	Minimum effective concentration	References
	rice	<i>Phaeocystis globosa</i>	chopped into different size: $\varphi = 2 \text{ cm}/\varphi > 0.3 \text{ mm}/\varphi < 0.3 \text{ mm}$	worked but concentration not record	[42]
	rice	<i>Microcystis aeruginosa</i>	straw rinsed with tap water, dried at 50°C for 3 days, cut, mortared and sieved through 1 mm	< 23.0 mg/L DW	[53]
	rice	<i>Syndra</i> , <i>Navicula</i> , <i>Nitzishia</i>	100 kg and 150 kg straw placed in two different ponds	NA	[74]
Cut and soak into water	barley	<i>Scenedesmus obliquus</i>	soaked into tap water at 21–25°C for different months	120.0 g/L WW for 3 months and 40 g/L WW for 4 months	[75]
	barley	<i>Klebsormidium rivulare</i> <i>Oedogonium</i> sp. <i>Spirogyra</i> sp. <i>Stigeoclonium tenue</i> <i>Ulothrix trentonense</i>	1 kg barley in 50 L water kept at at 20–25°C for 3 months	100.0 g/L WW	[34]
	barley	<i>Microcystis aeruginosa</i>	chopped and sieved (size 0.1–2.9 mm), extract filtered	0.5 g/L Liquor (aerated during bioassay)	[76]
	barley	<i>Microcystis aeruginosa</i> Sciento	2000 g straw soaked at room temp. for 3–6 months	70.0 mg/L DW	[77]
	barley	<i>Scenedesmus</i> sp., <i>Microcystis</i> sp.	fresh and decomposed barley straw powder boiled for 2 h and filtered to get extract	100.0 g/L straw (rotted)	[40]
	barley	<i>Heterocapsa pygmaea</i> , <i>Heterocapsa triquetra</i> , <i>Gyrodinium galatheanum</i> , <i>Gymnodinium sanguineum</i>	straw (2 cm), soaked in 18 L reverse osmosis (RO) water	36.0 g/LWW	[48]
	barley	<i>Microcystis</i> sp.	straw grinded, added to 100 mL distilled water	0.5 g/L WW	[78]
	barley	<i>Chlorella capsulata</i> , <i>isochrysis</i> sp., <i>Ankistrodesmus falcatus</i>	0.2 L water decanted into containers of dried barley straw for 4–7 days	1250.0 mg/L DW	[49]
	barley	<i>Microcystis aeruginosa</i> , <i>Dinobryon</i> sp., <i>Synura petersenii</i>	360 g straw chopped in 2 cm soaked in 18 L water, liquor obtained, incubated at 25°C for 60 days	20.0 g/L WW	[61]
	barley	<i>Ankistrodesmus falcatus</i> , <i>Coelastrum</i> sp.	25 g chopped straw soaked in 20 L aquarium water	1.3 g/L WW	[63]
barley	<i>Microcystis aeruginosa</i>	100 g straw (2 cm) in 5 L of water, decomposing straw incubated in water at room temperature	20.0 g/L WW	[43]	

(Continued)

Table 1 (continued).

Category	Straws type	Target algae	Straw pre-treatment method	Minimum effective concentration	References
barley		<i>Anabaena affinis</i> , <i>Microcystis aeruginosa</i> & <i>Scenedesmus quadricauda</i>	20 g of barley straw chopped by 2 cm into a distilled water soaked for 3 months	2.0 g/L WW	[79]
barley		<i>Microcystis aeruginosa</i>	straw cut into 2 cm, weighed & extracted by degradation in distilled water (2 g/L)	0.2 g/L straw extract	[80]
barley rice		<i>Phaeocystis globosa</i>	chopped into 2 cm pieces, sealed and soaked into distilled water at 22 ± 1°C for 72 h/1 month/3 months, filtered with 0.2 µm membrane before using	3.0 g/L extract for both autoclaved and not autoclaved groups	[42]
barley rice wheat		<i>Microcystis aeruginosa</i> FACHB-469	2 cm pieces soaked into tap water at 20–30°C for 1 month	0.2 g/L WW	[81]
rice		<i>Microcystis aeruginosa</i> <i>Oscillatoria</i> sp. <i>Anabaena azolla</i>	chopped into 2 cm pieces and soaked into tap water with aeration at 20–30°C for 1 month	5.0 g/L WW for <i>M. aeruginosa</i> and 15.0 g/L WW for <i>O. sp.</i> and <i>A. azolla</i>	[35]
rice		<i>Anabaena azolla</i> <i>Nostoc</i> sp.	chopped into 2 cm pieces soaked into tap water with aeration at 20–30°C for 1 month	5.0 ml/L for extract and 5 g/L for WW	[59]
rice		<i>Spirogyra</i> sp.	crushed, soaked into deionized water at 50°C for 2 days and filtered with 0.22 µm membrane to get extract, rotary evaporating the filtered extract to dry powder and re-dissolved into distill water	20.0 mg/L re-dissolved extract	[82]
rice		<i>Microcystis aeruginosa</i> FACHB-912	crushed and sieved over 40 mesh screen, soaked into distilled water at 25°C for 5 days, filtered with GF/C membrane before using	0.5 g/L extract for <i>M. aeruginosa</i> FACHB-912 and <i>A. flos-aquae</i> FACHB-245, 1.0 g/L for <i>S. capricornutum</i> FACHB-271, 2.0 g/L extract for <i>M. aeruginosa</i> FACHB-469 and <i>M. ichthyoblabe</i> FACHB-1294	[60]
rice		<i>Microcystis aeruginosa</i> 905	crushed and sieved over 5 mesh screen, autoclaved and soaked into deionized water at 4°C for 4 days	1.5 g/L WW	[38]
rice		<i>Microcystis aeruginosa</i> FACHB-912	chopped into 2 cm pieces soaked into distilled water at room temperature for 5 days, filtered with 0.45 µm membrane before using	3.0 g/L for extract and 2.0 g/L for WW	[83]

Table 1 (continued).

Category	Straws type	Target algae	Straw pre-treatment method	Minimum effective concentration	References
	rice	<i>Phaeocystis globosa</i>	chopped into 2 cm pieces, sealed and soaked into distilled water at 22°C in dark/light for 6 months, filtered with 0.2 µm membrane before using	4.1/4.2 g/L for 80% inhibition rate	[39]
	rice	<i>Microcystis aeruginosa</i>	crushed and sieved over 40 mesh screen, soaked into distilled water at 25°C for 5 days, filtered with GF/C membrane before using	0.5 g/L straw extract	[84]
	rice	<i>Scenedesmus</i> sp.	rice straw grinded, added to 100 mL distilled with 5 A filter paper	0.5 g/L straw extract	[78]
	rice	<i>Microcystis aeruginosa</i>	chopped into 2 cm pieces, sieved over 0.3/0.1 mm mesh screen	10.0 g/L DW for 2 cm pieces and 20.0 g/L DW for others	[85]
	rice	<i>Microcystis aeruginosa</i>	straw cut into 2 cm, sonicated for 10 min at 50°C extract concentrated to 1% dry wt	10.0 mg/L straw extract	[51]
	rice	<i>Microcystis aeruginosa</i>	straw crushed, sieved over 40 mesh screen, and soaked into BG11 solution at room temperature for 7 days	10.0 g/L straw extract	[54]
	rice	<i>Microcystis aeruginosa</i>	straw finely dried, transferred to beakers to decompose in ultrapure water	2.5 g/L WW	[86]
	wheat	<i>Microcystis aeruginosa</i>	8 g straw added to distilled water. Mixture autoclaved, solution centrifuged and supernatant used as leachates of wheat bran	1.6 g/L straw extract	[56]
	corn leaf and straw	<i>Alexandrium tamarense</i>	crushed and sieved over 160 mesh screen	1.0 g/L DW	[57]
	corn leaf	<i>Alexandrium tamarense</i> <i>Chattonella marina</i> <i>Heterosigma akashiwo</i>	crushed and sieved over 160 mesh screen	1.0 g/L DW	[62]
Chemical extraction	barley	<i>Microcystis aeruginosa</i> FACHB-912	2 cm pieces soaked into distilled water at for 15 days, filtered with 0.45 µm membrane and extracted with petroleum-ether, chloroform, ethyl acetate and <i>N</i> -butanol	74.6% inhibited for petroleum ether extract (5.0 ml/L) and 59.2% inhibited for ethyl acetate extract (5.0 ml/L) on 6th day	[87]

(Continued)

Table 1 (continued).

Category	Straws type	Target algae	Straw pre-treatment method	Minimum effective concentration	References
	barley	<i>Microcystis aeruginosa</i>	extracted chemical screened for solubility and reasonable concentrations from straw	> 10.0 µg/L straw extract	[88]
	barley	<i>Microcystis</i> sp.	straw dried at 70°C for 48 h, chopped in 1 cm extracts concentrated under vacuum at 42°C to yield extract	NA	[27]
Fungal degradation	barley	<i>Scenedesmus</i> sp.	autoclaved straw degraded by white rot fungi for 2 months at 20°C and 100% humidity	0.3 g/L degraded straw	[70]
Not mentioned	barley	<i>Microcystis aeruginosa</i> FACHB-315	degraded method not mentioned	10.0 ml/L straw extract	[37]
	barley rice	<i>Microcystis aeruginosa</i> <i>Chlorella vulgaris</i> <i>Scenedesmus obliquus</i>	degraded method not mentioned	0.2 g/LWW	[89]
	wheat	<i>Microcystis aeruginosa</i> <i>Chlorella vulgaris</i> <i>Scenedesmus obliquus</i> <i>Anabaena azolla</i>	degraded method not mentioned	0.2 g/L WW for <i>M. aeruginosa</i> <i>C. vulgaris</i> <i>S. obliquus</i> and 1.0 g/L degraded WW for <i>A. azolla</i>	[89]

DW: Dry weight, WW: Wet weight.

the grain, the crop species or their close relatives could be selected as fundamental ecological groups to inhibit blooms in brackish water, salt water and fresh water with poor flow. Integrating farmlands as regional wetlands is a key strategy, it may include the practice of restoring, creating or managing wetland habitats [97]. These selected crop types could be of agricultural straw origin that would act as riparian buffers, which removes pollutants, provide aquatic habitat and enhance in stream chemical process by releasing allelochemicals into the in situ water [97]. This ecological process could also help algal inhibition.

5 Mechanisms for the Algal Inhibition by Agricultural Straw

Present studies on the inhibition mechanisms mainly focus on the photosynthesis, membrane structure and enzyme activity of algal cells. And mechanism at genetic level was also concluded (Fig. 4).

5.1 Photosynthesis

Chlorophyll a plays an important role in energy capture and transfer during photosynthesis for microalgae [43,98]. The Chl a concentration is an indispensable index in photosynthetic rate determination. Recent investigations revealed that Chl-a decreased in algal cells treated by rice straw and barley straw [43,54]. Particularly, allelochemicals in barley straw, such as phenolic compounds [99] and salcolin [27] were found to reduce the Chl-a content in *M. aeruginosa* cells, hence, inhibit algal growth. Besides, Photosynthetic system II (PSII) of algal cells exposed to allelochemicals was widely explored using pulse amplitude modulated (PAM) fluorescence analysis [28,100,101]. The potential maximum photosynthetic capacity (Fv/Fm) of *M. aeruginosa* was inhibited by rice straw extracts, revealing that photosynthesis was its potential targeted pathway [55].

Table 2: Major anti-algal allelochemicals released from agricultural straw

Class	Allelochemicals	Source	Algae inhibited	References
Fatty acids	2-Methylbutanoic acid	barley	<i>Chlorella vulgaris</i>	[88]
	3-Methylbutanoic acid		<i>Microcystis aeruginosa</i>	
	Heptanoic acid			
	Hexanoic acid	barley	<i>Microcystis aeruginosa</i>	[88]
	Fumaric acid	rice	<i>Microcystis aeruginosa</i>	[95]
Simple aromatic compounds	Benzyl cyanide	barley	<i>Chlorella vulgaris</i>	[88]
	Acetophenone			
Phenols	<i>p</i> -Cresol	barley	<i>Chlorella vulgaris</i> <i>Microcystis aeruginosa</i>	[88]
	2,6-Dimethoxyphenol	barley	<i>Chlorella vulgaris</i>	[88]
	2-Phenylphenol	barley	<i>Chlorella vulgaris</i> <i>Microcystis aeruginosa</i> <i>Scenedesmus subspicatus</i>	[88]
Benzaldehydes	Benzaldehyde	barley	<i>Chlorella vulgaris</i> <i>Microcystis aeruginosa</i> <i>Scenedesmus subspicatus</i>	[88]
	Vanillin	barley	<i>Oscillatoria cf. chalybea</i>	[96]
	Syringaldehyde		<i>Selenastrum capricornutum</i> <i>Anabaena</i> sp. LP 691 <i>Pediastrum simplex</i>	
Benzoic acids	Benzoic acid	barley	<i>Chlorella vulgaris</i> <i>Microcystis aeruginosa</i>	[88,95]
	<i>p</i> -Hydroxybenzoic acid	barley	<i>Oscillatoria cf. chalybea</i> <i>Selenastrum capricornutum</i> <i>Anabaena</i> sp. LP 691 <i>Pediastrum simplex</i>	[96]
	Salicylic acid	rice	<i>Microcystis aeruginosa</i>	[95]
	Syringic acid	barley/rice	<i>Microcystis aeruginosa</i>	[95,96]
	Vanillic acid		<i>Oscillatoria cf. chalybea</i> <i>Selenastrum capricornutum</i> <i>Anabaena</i> sp. LP 691 <i>Pediastrum simplex</i>	
Phenylpropanoids	<i>trans</i> -Cinnamic acid	barley	<i>Oscillatoria cf. chalybea</i> <i>Selenastrum capricornutum</i> <i>Chlorella vulgaris</i>	[88,96]
	<i>p</i> -Coumaric acid	barley	<i>Microcystis aeruginosa</i>	[95]
	Caffeic acid Chlorogenic acid	barley	<i>Oscillatoria cf. chalybea</i>	[96]
	Sinapic acid		<i>Selenastrum capricornutum</i>	
	<i>trans</i> -Ferulic acid	barley	<i>Oscillatoria cf. chalybea</i> <i>Selenastrum capricornutum</i> <i>Anabaena</i> sp. LP 691 <i>Pediastrum simplex</i>	[96]
Flavonolignans	Salcolin A Salcolin B	barley	<i>Microcystis aeruginosa</i>	[27]
Others	Tannic acid	barley	<i>Oscillatoria cf. chalybea</i> <i>Selenastrum capricornutum</i>	[96]

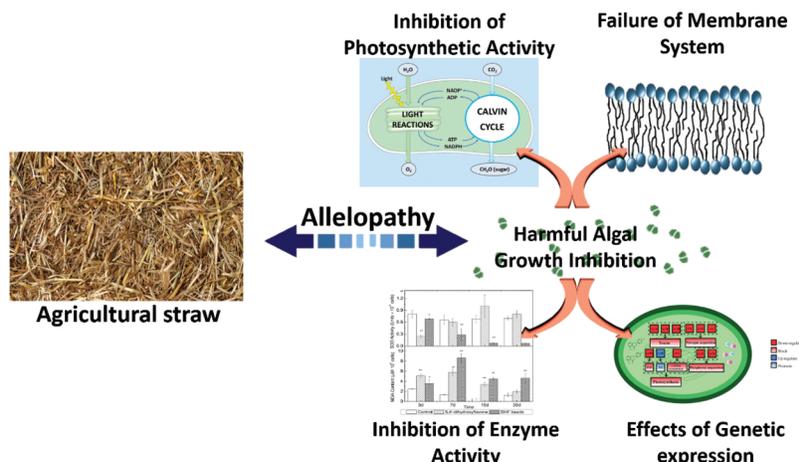


Figure 4: Mechanism pathway of the agricultural straw inhibition on phytoplankton

5.2 Cell Membrane and Morphological Changes

The mode of action of algae response to antialgal chemicals can be divided into; ‘algicide’ (killing algae) and ‘inhibitor’ (preventing algae growth). Cell membrane integrity was disrupted when the allelochemicals acted as ‘algicide’. For instance, salcolin A and salcolin B in barley straw were ‘algistatic’ and ‘algicidal’ respectively to *M. aeruginosa* [27]. Recent research demonstrated that the aqueous extracts of rice straw accelerated the lysis and crack of *M. aeruginosa* cells, and lead to cell death [41,54,82]. At low exposure concentrations, the *M. aeruginosa* cells shrunk [60]. When exposure concentration increased, *M. aeruginosa* cell swelled. Corn leaves were found to split group *Alexandrium tamarense* cells into unicellular, make cells losing the ability to swim, swollen cell protoplast and finally cells ruptured [62]. It is important to explore the effect of agricultural straw on algae cell structure as the cell membrane was always influenced by antialgal chemicals leading to intracellular microenvironment homeostasis disruption [102].

5.3 Enzyme Activity

Enzyme plays an important role in organisms. *Stratiotes abides* extracts reduced the alkaline phosphatase activity (APA) of planktonic cyanobacterium *Anabaena variabilis*. The microcystin synthesis related enzymes were enhanced and the toxin production were promoted in epiphytic cyanobacteria *Merismopedia tenuissima* and *Leptolyngbya boryana* [103]. Environmental stress could cause high cellular reactive oxygen species (ROS) levels that affect the antioxidant enzyme activities (i.e., Superoxide Dismutase, Malondialdehyde and Catalase) of algae cells [104,105]. High cellular ROS levels was harmful to DNA, RNA, proteins and lipids and also acted as a signal to trigger programmed cell death (PCD) [106,107]. The wheat bran leachate (WBL) could increase the intracellular ROS levels of *M. aeruginosa* significantly at low concentration (0.8 g/L), indicating that oxidative damage contributed to the algal inhibition [56]. Barley straw extracts suppressed the growth of algae by the way of promoting intracellular ROS levels to reduce enzyme activity [43]. Rice straw extracts were also observed to alter SOD activities [41].

5.4 Genetic Expression

Genetic expression regulated the growth of cell initially and metabolic pathway of algae was disrupted successively, even induced to programmed cell death (PCD) under external stress [108,109]. The genes related to photosynthesis (incl. *psbA*, *psbD1*, *psaB*, *rbcL*), toxin synthesis (incl. *mcyA*, *mcyB*, *mcyD*, *mcyH*), nutrient utility (incl. *nirA*, *ntcA*, *amt*, *phoU*, *pstA*), antioxidant protein peroxiredoxin synthesis

(incl. *prx*), fatty acid synthesis (incl. *fabZ*) and repair of biological macromolecules (incl. *recA*, *grpE*) were widely studied in *M. aeruginosa* [110–112]. For instance, Shao et al. found that WBL down-regulated expression of *psbA*, *fabZ* and *prx* genes, and up-regulated *mcyB* expression [56]. Recently, transcriptomics was applied to investigate the changes of gene transcription of harmful algae under external stress (i.e., nutrient limitation, predators) [113]. However, research on transcriptional responses of algae to allelochemicals was still lacking [114].

6 Challenges and Field Applications of Using Agricultural Straws as Anti-algal Agents

Our investigation reveals that about 92% of these experiments are done in laboratory while the remaining 8% are carried out in rivers, reservoirs, lakes, canal and ponds (Fig. 5). More field application is needed to overcome this challenge. The lack of a thorough clarification and summary of specific functional mechanisms of allelochemicals has constrained our understanding of its ecological significance. Researches that have been carried out have not been able to prove the specificity of each of these allelochemicals to specific algal strains so as to provide a data-base clear information, this is clearly a challenge to the science community. However, this review seeks to encourage researches that would illustrate the general characteristics of algae, study their physiology and allelopathic pathways of these allelochemicals. Through this approach, allelochemicals that are species-specific could be produced. This approach to suppress HABs could decrease the use of harmful chemical herbicides while providing an alternative sustainable technology of terrestrial crop management in the future.

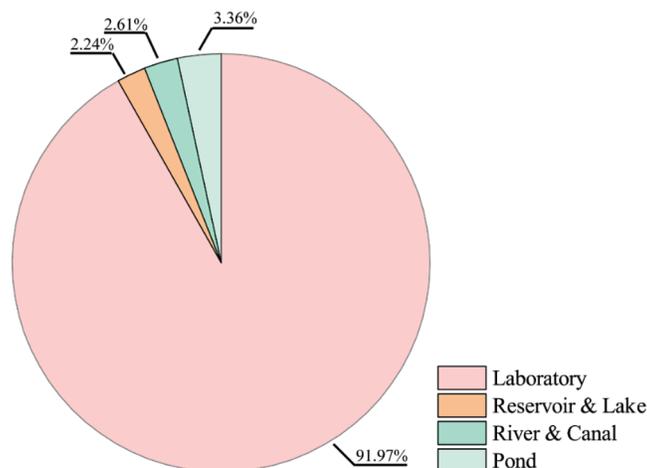


Figure 5: Percentages of the HABs control application of agricultural straws in various types of water body, including laboratory, reservoir & lake, river & canal and pond

6.1 Advantages of Using Agricultural Straw Extracts

One of the good qualities of agricultural straw extracts used for the control of cyanobacterial blooms [99], is its ability to be easily degraded in the environment which it is used. With better pre-treatment methodology using white-rot fungi to improve lignin degradation in straw, thereby, improving algal inhibition [70], it is a step in the right direction. In most cases, the allelopathic compounds found in these straws show inhibitory effects on bloom forming cyanobacteria and other algal species. As stated earlier, agricultural straw is sometimes burnt directly, which constitute an emission problem, some farmers inhume them and generally take them as waste products. These materials are cheap, easy to come by and above all it does not require a complex process to extract. Lastly, none of the reviewed works have reported any strong negative impacts on both aquatic animals and human [32,61,115,116], it is therefore

safe to conclude that agricultural straw application on the inhibition of algal bloom is safe, effective, eco-friendly and cheap. For instance, there are already some commercial products made of barley straw, including bales (Swell UK, UK), liquid (King British, UK) and balls (Gardening-naturally, UK) (Fig. 6). These products are mostly designed for pond use.



Figure 6: Commercial products from barley straw for algal control

6.2 Disadvantages

The major disadvantages occur during direct use of straw on the field, including taking a longer time to biodegrade due to low temperature [117], blocking river channels and constituting nuisance to the aquatic environment may also occur. Uncontrolled decomposition may cause secondary pollution [93,118], introduce excessive nutrients into water, and bring the risk of microbial/viral attached on straw surface to the aquatic ecosystem.

6.3 Recommendations

- (i). Most reports show that the allelopathic effect of agricultural straws are algistatic and not algicidal, therefore we recommend that the application approach should be proactive. It should be applied at early algal bloom stage rather than at full bloom condition.
- (ii). In view of the fact that agricultural straws have high potential of being effective and environmentally safe agents for the control of cyanobacterial blooms and other nuisance algae. It is pertinent to continue to isolate and discover allelochemicals with anti-cyanobacterial or anti-algal properties and optimize its application, i.e., slow release beads carrying the allelochemicals [93]. Species-specific allelochemical mechanism should be properly investigated so as to broaden the scope of effective inhibition.
- (iii). A commercialized method of allelochemical production should be encouraged and a risk evaluation be properly documented before marketing.
- (iv). A system of combining straw extract application with other approaches, such as bio manipulations (introducing some plants that can absorb excess nutrients and introducing some organisms that feed directly on algae is also recommended), enforcing strict regulations on the use of fertilizer and proper waste treatment by industries.
- (v). It is a known fact that some crops contain allelochemicals capable of inhibiting algae, these allelopathic compounds could also be released naturally into water bodies. We therefore recommend that a system of farming where barley, rice and other related crops be planted in the upstream catchment of water bodies, i.e., wetland areas. These crops will absorb nitrates and phosphate, reduce eutrophication, release allelochemicals to inhibit algal blooms and also serve as food for man. In this way, a novel way of solving multiple problems using an eco-friendly method could be achieved.

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